

Positive-Ionic Mobility and a Hierarchy of Ions in Normal Liquid ^3He

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We have measured positive-ion mobilities in liquid ^3He down to 3 mK. At 5 bar the low-temperature mobility is given by $\mu = (0.17 - 4.41 \ln T) \times 10^{-6} \text{ m}^2/\text{V} \cdot \text{s}$. At 27 bar two mobilities are seen, $\mu_{\text{fast}} = (-12.85 - 6.21 \ln T) \times 10^{-6} \text{ m}^2/\text{V} \cdot \text{s}$ and $\mu_{\text{slow}} = (-6.26 - 3.64 \ln T) \times 10^{-6} \text{ m}^2/\text{V} \cdot \text{s}$. At temperatures near the discontinuity we see multiple mobilities and time-dependent conversion from one species of ion to another.

We have measured positive-ion mobilities, μ_+ , in liquid ^3He down to 3 mK at several pressures. We have been able to observe several distinct and different species of ions simultaneously. At the temperature of the discontinuity in μ_+ observed in earlier measurements^{1,2} at around 50 to 100 mK we see a transition from one dominant ion type to another with several intermediate states.

Measurements are made in an ion cell similar to that used by us earlier.³ The electrode structure is shown in the inset in Fig. 1. The free volume of liquid in the cell is in thermal contact with a cooling salt of cerium magnesium nitrate also used as the thermometer. Ions are pro-

duced by transiently pulsing the field-emission tip above its positive-emission potential. By suitable manipulation of the potentials at the gate (grids 1 and 2), the pulse can be time marked and the transit time across the uniform-field drift space determined by the arrival of the pulse at the Frisch grid, grid 3, at which point it is sensed by the collector. The collector output is fed to a signal averager. The mean mobility, $\mu = \text{velocity/field}$, can then be obtained directly. The mobility obtained in this way becomes field dependent at the lower temperature because of the increasing nonlinearity in the velocity-field relation. This has the result that a measurement of velocity/field at finite field falls below μ_0 (velocity/field at zero field) with decreasing temperature and finally saturates at a constant value. Therefore we have chosen to measure μ differentially. The transit time, τ , is measured for a number of drift field voltages, V_D , (usually nine) from 90 to 2000 V/cm. At the lower fields the data all fall on a straight line on a plot of τ vs $1/V_D$, i.e., $\tau = L^2/\mu V_D + \text{const}$ (L being the length of the drift space), and thus the slope taken from such a plot yields μ directly.

In the first run with the cell we measured μ_+ at 5 bar down to 2.5 mK using conventional gating techniques. The results were almost identical to those of Roach, Ketterson, and Roach² measured at 60 Torr, showing a discontinuity at ~ 50 mK and a mobility gradually leveling off to a constant value below about 8 mK. Next we pressurized to 27 bar and repeated the measurement but now at low temperatures we could see two pulses of ions, of similar amplitude, arriving at the collector. Under the influence of Bowley's ideas⁴ that positive ions in ^3He may attract a layer of ^4He from impurities in the liquid at higher temperatures, we made a number of plots of τ vs $1/V_D$ to see if the two species of ions were created simultaneously at the tip or if one was created out of the other. At low temperatures the two were found

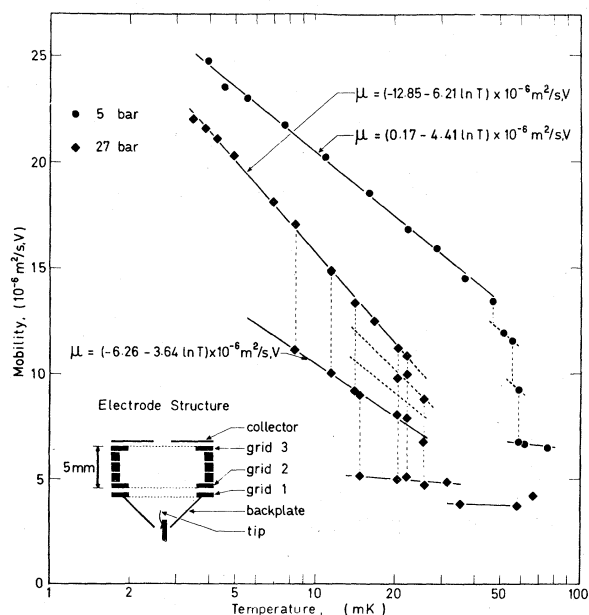


FIG. 1. Positive-ion mobility in liquid ^3He . Fine vertical dashed lines join mobilities seen simultaneously at one temperature. Heavy dashed lines indicate estimated mobilities where we can detect the ion pulses but cannot follow them over a large enough range of V_D to define μ .

to be created simultaneously on the time scale of our measurements.

We then reduced the pressure to 23 bar. We still had two pulses at low temperatures but the slower one was of much lower amplitude than at 27 bar. At the discontinuity we studied the behavior with several plots of τ vs $1/V_D$ and saw definite signs of multiple mobilities and evidence of conversion from one species of ion to another with increased transit time. On repressurizing to 29 bar, in an attempt to increase the amplitude of the slow pulses, we found to our surprise no slow pulses at all. It was during this run with the appearance of very large discrepancies between the value of μ deduced from the plots of τ vs $1/V_D$ and that of the straightforward gating method that we realized that the latter technique was measuring a field-dependent quantity and we went over to using plots of τ vs $1/V_D$ exclusively to determine μ . We now found that the low-field mobility, instead of leveling off, was linear in $\ln(1/T)$ down to the lowest temperatures.

We had earlier concluded that μ_{slow} might be due to solid ^4He ions emitted at regions of the tip coated with an electrostricted layer of residual solid ^4He rather than solid ^3He . Since there are no states for ^4He atoms in the liquid at low temperatures, such ions would be very stable. Now, however, we had no μ_{slow} , but at this point the liquid in the cell had always been below ~ 100 mK and at or above 23 bar for three weeks and we guessed that the cumulative effect of pulsing the tip had been to clean off the ^4He , since there would be no mechanism for replenishing it if any ^4He film were solid at these pressures. If this were so, then a run to remeasure the 5-bar data would render any film mobile and a subsequently return to high pressure should bring back μ_{slow} which gratifyingly proved to be the case.

During the final run looking at both μ_{fast} and μ_{slow} at 27 bar we ran the tip at a very low voltage and realized that we could see *five* distinct pulses simultaneously. These consisted of μ_{fast} , two intermediate mobilities which we had earlier seen only transiently at the discontinuity, μ_{slow} , and a very slow mobility corresponding to the almost temperature-independent value seen above the discontinuity.

Figure 1 shows the results for 5 and 27 bar with the data taken as described above and μ deduced from the expression $\mu = L^2 d(1/V_D)/d\tau$, where L is the length of the drift space, τ the transit time (taken arbitrarily from the initiation of the tip pulse), and V_D the voltage drop across

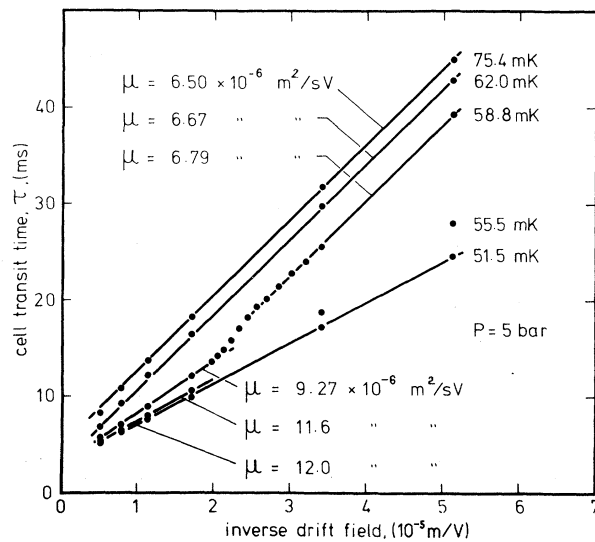


FIG. 2. The high-field part of the plots of τ vs $1/V_D$ for the 5-bar data near the discontinuity.

the drift space.

The 5-bar data taken at low drift fields show a single-valued mobility below the region of the discontinuity (although we have not yet searched here with low tip potential) which fits the expression $\mu = (0.17 - 4.41 \ln T) \times 10^{-6} \text{ m}^2/\text{s} \cdot \text{V}$ and shows no sign of the leveling off at low temperatures seen earlier. As we reach temperatures near the discontinuity we see the first evidence of multiple mobilities with two intermediate-mobility "states" between the low-temperature and high-temperature curves. At several temperatures we see two mobilities simultaneously and we see the slower species of ion being created out of the faster one, the conversion being more complete the greater the transit time. Figure 2 shows the high-field part of the plots of τ vs $1/V_D$ for several temperatures near the discontinuity. The kink in the 58.8-mK data, representing such conversions, is quite clear. Further, on extrapolation, the low-mobility section of this curve intersects the τ axis at negative time showing that the ions reaching the collector with that mobility have not had time to traverse the whole cell (i.e., they must have started out faster). The successive displacement of the two higher-temperature curves to the left indicates a faster conversion and also implies that a higher mobility is also present at these temperatures were we to work at fields high enough to measure it.

The 27-bar data look very different. At low temperatures there are two mobilities which fit

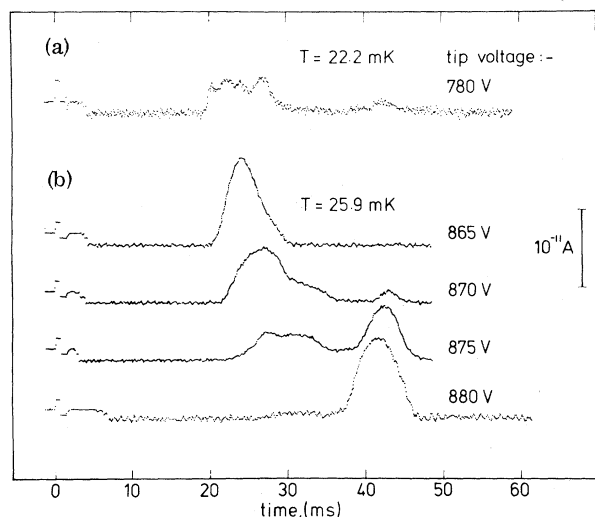


FIG. 3. (a) A typical averaged collector signal at 27 bar showing multiple pulses at low tip potential. (b) Several collector signals at 27 bar near the discontinuity showing the sensitivity of the various pulses to the tip potential. The first vertical step in each trace marks the beginning of the tip pulse.

the expressions $\mu_{fast} = (-12.85 - 6.21 \ln T) \times 10^{-6} \text{ m}^2/\text{s} \cdot \text{V}$ and $\mu_{slow} = (-6.26 - 3.64 \ln T) \times 10^{-6} \text{ m}^2/\text{s} \cdot \text{V}$. The figure shows μ_{slow} no lower than 8 mK only because we have no ungated data below this. Whether or not μ_{slow} is present depends on the pressure and temperature history. If the emission tip potential is reduced to a very low value, unfortunately only tried at 15 mK and above, then two further intermediate pulses appear between μ_{fast} and μ_{slow} and a mobility very similar to the high-temperature constant value also appears. A typical signal is shown in Fig. 3(a). At the discontinuity, which of these mobilities dominates is a very sensitive function of the tip potential; see Fig. 3(b). A few millikelvins above the discontinuity there also appears to be a small jump from one constant value to a lower one, a similar effect being seen in the 23-bar data.

We also have data over the whole range for 29 bar (without μ_{slow} because of tip cleaning) and around the discontinuity for 23 bar, but neither taken at low tip voltage. The results are qualitatively similar to those at 27 bar taken in the same way.

How are we to interpret these results? The conventional picture of the positive ions as a snowball of solid ^3He electrostricted around the ionized atom can only be valid in the complete absence of ^4He contamination. Our ^3He contains

about 5×10^{-4} parts of ^4He which is of the correct order of magnitude to form a film. It is clear that we observe several discrete states of the ions which presumably represent states of greater or lesser ^4He admixture. Certainly at the very low temperatures and high pressures, where we can clean enough ^4He from the tip region to suppress μ_{slow} , then the presence of μ_{slow} must be due to ^4He , possibly solid on the tip, but it does not follow that μ_{fast} is necessarily a ^3He snowball in ^3He . At higher temperatures the temperature-independent μ seen above the discontinuity may be due, as Bowley has suggested,⁴ to ^3He ions completely glazed with ^4He . Nevertheless, how the several different mobilities seen near the discontinuity fit into this picture is not easy to see. That they are influenced by the tip voltage may be because near the tip the local temperature of the liquid may be rather high and thus will contain a higher concentration of dissolved ^4He . Changing the tip voltage will tend to expand or contract this heated region and influence the local impurity level.

It is interesting that the slope of the low-temperature mobility at 5 bar is closer to that of μ_{slow} than that of μ_{fast} for the 27-bar data. This may mean that at pressures below the ^4He film melting pressure the mobility may always be influenced by ^4He until the sample is of such high purity that no film is possible. We have gated data for 5, 11, and 20 bar from an earlier cell⁵ which show that just below the discontinuity (where gated and differential measurements are not significantly different) the mobilities for the three pressures have similar temperature dependencies so that there may well be a jump in the slope of the fastest μ between 20 and 27 bar.

In conclusion we can say that positive-ion mobilities which we can follow over a significant temperature range are all linear in $\ln(1/T)$ and also that there are several discrete ionic states possible in liquid ^3He .

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¹A. C. Anderson, M. Kuchnir, and J. C. Wheatley, Phys. Rev. **168**, 261 (1968).

²P. D. Roach, J. B. Ketterson, and P. R. Roach, Phys. Rev. Lett. **39**, 626 (1977).

³C. N. Barber, P. V. E. McClintock, I. E. Miller, and G. R. Pickett, *Phys. Lett.* **54A**, 241 (1975), and references therein.

⁴R. M. Bowley, to be published.

⁵C. N. Barber, thesis, University of Lancaster (to be published).

Temperature Anomalies of Hyperfine Fields of *s-p* Impurity Elements in Cobalt

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The hyperfine magnetic field H_{hf} at extremely dilute impurities of Ga, Ge, and As in a single crystal of hcp Co has been measured as a function of the temperature T up to $T \cong 900^\circ\text{K}$. The functions $H_{\text{hf}}(T)$ exhibit dramatic differences for these adjoining elements; while $H_{\text{hf}}(T)$ in AsCo scales with the hcp host magnetization $\sigma(T)$, $H_{\text{hf}}(T)$ in GaCo decreases much faster than $\sigma(T)$ and $H_{\text{hf}}(T)$ in GeCo *increases* by $\sim 25\%$. This is the first observation of an increase with temperature of the magnitude of the hyperfine field at an impurity atom in any ferromagnetic metal.

It is well known that the systematics of hyperfine fields H_{hf} at dilute nonmagnetic impurities in 3d ferromagnets exhibit striking variations with the impurity charge number Z . The sign of H_{hf} reverses from negative to positive regularly at the middle of the *s-p* series of each period, e.g., near Ge (4*sp*) and near Sn (5*sp*). Although these systematics have been studied for over a decade, a complete understanding is still lacking. It is believed that these sign reversals of H_{hf} result from competition between conduction-electron polarization (CEP) and overlap-exchange polarization of the valence *s*-like electrons of the impurity, but the physical origins of these contributions, especially the latter, remain controversial.¹ Direct experimental information on the components that make up H_{hf} is scarce and not easy to obtain. From this point of view, the impurities immediately around the point of sign crossover of H_{hf} (e.g., Ge or Sn) for which the different contributions compete nearly equally and produce a small net field are particularly interesting. In these cases, differences between the temperature dependence of those components sensitive to radial dependence or volume effects, and that of the CEP which is expected to scale roughly as the host magnetization $\sigma(T)$, could produce large effects in the observed $H_{\text{hf}}(T)$. Indirectly, this could lead to important insight into the origin of these hyperfine fields.

Experimental data on $H_{\text{hf}}(T)$ are available for *sp* impurities, mainly for an Fe host.² These

show that although small deviations of the order of several percent from the reduced magnetization of the host $\sigma(T)/\sigma(0)$ are commonly observed, it is not possible to draw clear conclusions on the origin of these effects. On the other hand, $H_{\text{hf}}(T)$ for Sn in Co, which is located at the sign crossover of H_{hf} of the 5*sp*-series impurities, is known to be strikingly anomalous.³ This anomaly is thought to be due to the influence of thermal vibrations on the overlap contribution to H_{hf} .³ This is, however, not completely established and furthermore the SnCo anomaly remains so far the only one of its kind. It is therefore interesting to investigate whether it is an isolated case or whether similar anomalies are present systematically for the isoelectronic 4*sp* impurities in a Co host.

In this Letter, we report measurements of $H_{\text{hf}}(T)$ in the range $T = 77$ to 900°K for Ga, Ge, and As impurities (concentration $\sim 10^{-10}$) in a single crystal of Co. Our results show that the temperature dependences of the hyperfine field at these adjacent impurities are dramatically different. For As, $H_{\text{hf}}(T)$ follows closely the host magnetization $\sigma(T)$ in the hcp phase; for Ga, $H_{\text{hf}}(T)$ decreases much more rapidly than $\sigma(T)$, while for Ge, $H_{\text{hf}}(T)$ displays a deviation from $\sigma(T)$ of the opposite kind, viz., it actually *increases* by about 25% in the temperature range studied. Considering that the host magnetization *decreases* by about 6% in this range, the GeCo result is remarkable and represents the first